

No new matter has been added by the above amendment. Claims 1, 2 and 4-13 are now pending in the application.

REMARKS

The rejections of Claims 1, 2 and 4 under 35 U.S.C. § 102(b) as anticipated by EP 792858 (Takagawa et al), and of Claims 3 and 5 under 35 U.S.C. § 103(a) as unpatentable over Takagawa et al in view of U.S. 6,057,487 (Munson et al), are respectively traversed.

In view of the above-discussed amendment, which, *inter alia*, incorporates the subject matter of Claim 3 into Claim 1, the anticipation rejection is now moot. Accordingly, it is respectfully requested that it be withdrawn.

The present invention relates to a process which, on the basis of suitable crystallization and filtration techniques, allows the separation of 2,6-dimethylnaphthalene (2,6-DMN) from mixtures in which it is present with isomers thereof.

As described in the specification beginning at page 1, line 15, 2,6-DMN has been synthesized by many different processes in the prior art, often resulting as part of a mixture containing isomers thereof and other compounds. Various techniques have been used to separate 2,6-DMN from such mixtures. Such techniques have been problematical. Two phenomena complicate the separation of high purity 2,6-DMN: one, the crystal obtained by crystallization from the molten state has such a morphology that, after separation of the mother liquor by filtration, the residual wetting of the solid is high and therefore the titer of 2,6-DMN in the solid is low; and two, the presence of a co-crystallization phenomenon due to the presence of other compounds in the solid, and particularly, 2,7-DMN. For example, the 2,7/2,6 ratio in the solid is about 10% of this ratio in the mother liquor. Solutions to this problem have been suggested in the prior art, but these are also problematical.

The present invention addresses the problems of the prior art and is surprisingly simple and effective, and results in the production of 2,6-DMN having a very high degree of purity, with contemporary recoveries close to the theoretical value, without any of the limits and disadvantages which characterize the methods described in the state-of-the-art.

As recited in above-amended Claim 1, the present invention is a process for the separation of 2,6-dimethylnaphthalene from a starting mixture containing it and isomers thereof comprising the following operations: (A) crystallization of 2,6-dimethylnaphthalene by the addition of a solvent and cooling of the mixture thus obtained to a temperature higher than the highest formation value of any eutectic of 2,6-dimethylnaphthalene and another isomer in the mixture, whereby a mother liquor containing solid, said solid comprising 2,6-dimethylnaphthalene, is produced; (B) removal of the mother liquor by repeated washings with a solvent; (C) dissolution in a solvent of the solid obtained, whereby a solution is produced; (D) crystallization of said solution by cooling, whereby a suspension is produced; (E) filtration of said suspension, whereby said 2,6-dimethylnaphthalene is separated, and wherein the solvent used for one or more of operations (A), (B) and (C), is selected from the group consisting of low molecular weight aliphatic alcohols, glycols, and mixtures thereof.

In a preferred embodiment, the same solvent is used for each of operations (A), (B) and (C), and more preferably, is methanol. When an aliphatic alcohol is used as the solvent, the above-discussed co-crystallization is substantially reduced in degree and consequently, the isomers present as solid in the 2,6-DMN crystal are in a lower amount. For example, the 2,7/2,6 ratio in the solid is about 2% of the 2,7/2,6 ratio in the mother liquor.

Takagawa et al, which was cited in the corresponding European Search Report as "A", i.e., technological background, category only, discloses a process for the production of highly pure 2,6-DMN by crystallization from a mixture of DMN isomers. Takagawa et al

disclose aliphatic saturated hydrocarbons and alicyclic saturated hydrocarbons only as the solvent for use in their process (page 5, lines 19-20). Takagawa et al require various compositional limitations in their starting mixture (page 6, line 19 ff), particularly that it contain at most 10% by weight of 2,7-DMN (page 7, lines 10-12). Indeed, the examples in Takagawa et al confirm that their process is applicable only with relatively small amounts of 2,7-DMN. Either the amount of 2,7 isomer is so small as to not even be quantified or, as in Examples 5 and 6, it is less than 5 wt%. The present invention, on the other hand, is not so limited and indeed, the Example herein, described beginning in the specification at page 10, line 10, uses a starting mixture having a concentration of 2,7-DMN of 15.9 wt%. (A)

While Applicants appreciate that the presently-claimed process is not limited to one requiring a minimum amount of 2,7-DMN greater than the 10 wt% 2,7-DMN maximum of Takagawa et al, nevertheless, this difference highlights the fact that beside the difference in solvents, the presently-claimed invention is otherwise different from the crystallization process of Takagawa et al. Note that the only limitation herein with regard to a starting mixture containing 2,7-DMN isomer is that the 2,6 isomer be present in an amount higher than that of its eutectic with the other isomers. || ?

The Examiner erroneously finds that Takagawa et al's process involves crystallization at a temperature of ²25°C. Rather, while the temperature may be 25°C, it is not required to be. Indeed, Takagawa et al disclose that the crystallizing operation temperature is preferably 10-60°C, and is correlated with the amount of solvent to be used, the solubility of 2,6-DMN in the solvent, the concentration of 2,6-DMN in the mixture of DMN isomers, and the concentration of the suspension at the end of the crystallization (page 7, lines 23-25). The value of 25°C in the description at page 7, lines 29-53, and relied on by the Examiner, is thus based on the specific composition of isomers disclosed therein and the use of heptane as the (2)

solvent. In addition, the temperature was not selected to obtain the maximum crystallization yield of 2,6-DMN, but to obtain a crystallization slurry having characteristics of ease of handling for transportation or stirring. In other words, the description of a temperature in Takagawa et al of 25°C is meaningless with regard to the presently-recited solvent. ✓

In support of the above, a mathematical model was used to compare the results obtained in Example 1 of Takagawa et al and results obtainable using the same crystallization temperature and the same isomer mixtures, but with methanol used as the solvent. The mathematical model simulated the crystallization at 20°C of the Takagawa et al's Example 1, carried out in heptane, and in methanol, as shown in Tables A and B, respectively, **submitted herewith**. By comparing the results in Tables A and B, it can be seen that the crystallization yield of 2,6 DMN significantly increases when changing from heptane (66.4%) to methanol (80.9%). In addition, the purity in the panel remains practically unchanged, i.e., 99.31% using heptane, and 99.34% using methanol. (v)

Notwithstanding all that has been discussed above, Takagawa et al neither disclose nor suggest the presently-recited combination of steps (A)-(E) of Claim 1 herein, notwithstanding differences in solvent. ✓

Munson et al is drawn to a method for producing 2,6-DMN from mixed dimethylnaphthalenes by crystallization, adsorption and isomerization. Particularly, Munson et al disclose a method of purifying 2,6-DMN from a feed mixture of DMN isomers and near-boiling compounds comprising the steps of crystallizing the mixture to precipitate a eutectic composition comprising 2,6-DMN and 2,7-DMN; optionally dissolving the eutectic composition in a solvent; and recovering a predominantly 2,6-DMN composition from the dissolved eutectic composition by adsorbing out non-2,6-DMNs onto an adsorption column (column 3, lines 20-30). As a means for separating the 2,6-DMN/2,7-DMN eutectic, Munson

et al disclose solvent crystallization using a solvent which includes, *inter alia*, the hydrocarbon solvents of Takagawa et al and the presently-recited alcohols (column 5, line 49 ff).

The Examiner finds that in view of the above-discussed disclosure with regard to solvents, it would have been obvious to one of ordinary skill in the art to substitute the hydrocarbon solvent of Takagawa et al with the alcohol solvent of Munson et al. However, this finding ignores essential differences between Takagawa et al and Munson et al, such that one skilled in the art would not have made this substitution, for reasons now discussed.

In Takagawa et al, the initial crystallization of 2,6-DMN is with the use of a solvent. In Munson et al, it is disclosed that crystallization of the eutectic be carried out in the absence of a solvent when the concentration of 2,6-DMN and 2,7-DMN isomers is higher than 20% and more preferably higher than 90% (column 5, lines 29-32), while a solvent should be used only when the concentration of 2,6-DMN and 2,7-DMN isomers is low (column 5, lines 49-52). Thus, Munson et al suggests that a DMN starting mixture of the type disclosed in Takagawa et al (when the concentration of 2,6-DMN and 2,7-DMN isomers is higher than 20%) should be subjected to a melt crystallization, i.e., a crystallization without a solvent. In other words, the disclosure of solvents in Munson et al would not be considered to be relevant to the DMN compositional starting mixtures of Takagawa et al. 3

Moreover, even if Takagawa et al and Munson et al were combined, the result would not be the presently-claimed invention, because the present invention does not seek to precipitate a eutectic composition comprising 2,6-DMN and 2,7-DMN. Indeed, step (A) of Claim 1 crystallizes down to a temperature which is higher than the formation value of such a eutectic.

In addition, while Munson et al is concerned with starting mixtures having a significant amount of 2,7-DMN with regard to 2,6-DMN, Takagawa et al relates more to starting mixtures containing very little relative amount of 2,7-DMN, as discussed above.

Further, Munson et al requires the use of an adsorbent to separate 2,6-DMN from 2,7-DMN. On the contrary, the present invention does not involve the use of adsorbents.

Finally, it is noted that Example 4 of Munson et al, which uses meta-xylene as a crystallization solvent, produces a crystallization yield of 16.4%, which is significantly lower than that obtainable by the presently-claimed invention.

For all the above reasons, it is respectfully requested that the rejections over prior art be withdrawn.

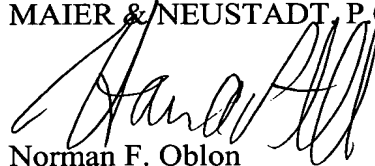
The rejection of Claims 1-5 under 35 U.S.C. § 112, second paragraph, is respectfully traversed. Indeed, the rejection is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that it be withdrawn.

The objection to Claims 3-5 as being in improper multiple dependent form is now moot in view of the above-discussed amendment. Accordingly, it is respectfully requested that it be withdrawn.

All of the presently pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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IN THE CLAIMS

--1. (Amended) A process for the separation of 2,6-dimethylnaphthalene from [mixtures] a starting mixture containing it and isomers thereof comprising the following operations:

[-] (A) crystallization of 2,6-dimethylnaphthalene by the addition of a solvent and cooling of the mixture thus obtained to a temperature higher than the highest formation value of [the first] any eutectic of 2,6-dimethylnaphthalene and another isomer in the mixture, whereby a mother liquor containing solid, said solid comprising 2,6-dimethylnaphthalene, is produced;

[-] (B) removal of the mother liquor by repeated washings with a solvent;

[-] (C) dissolution in a solvent of the solid obtained, whereby a solution is produced;

[-] (D) crystallization of said solution by cooling, whereby a suspension is produced;

[-] (E) filtration of said suspension, whereby said 2,6-dimethylnaphthalene is separated, and

wherein the solvent used for one or more of operations (A), (B) and (C), is selected from the group consisting of low molecular weight aliphatic alcohols, glycols, and mixtures thereof.

2. (Amended) The process for the separation of 2,6-dimethylnaphthalene according to [the previous claim] Claim 1, wherein the starting mixture contains [the same] 2,6-

dimethylnaphthalene in a concentration higher than [the] its eutectic concentration [ratios] with [the other] isomers thereof that are present in said starting mixture.

3. (Cancelled).

4. (Amended) The process for the separation of 2,6-dimethylnaphthalene according to [the previous claim] Claim 1, wherein the same solvent is used for [the various] each of operations (A), (B) and (C).

5. (Amended) The process for the separation of 2,6-dimethylnaphthalene according to [the previous claim] Claim 4, wherein the solvent used is [preferably] methanol.

6-13. (New).--

TABLE A

Preparation of 2,6 Dimethylnaphthalene having a high purity by crystallization with n-heptane
Example 1 of EP 0 792858

CRYSTALLIZATION CARRIED OUT AT: 20 °C

	Initial charge		% N-heptane		Sol. 2,6 DMN		Mothers		Solid		Wet/Solid		Panel	
	g	% DMN	g	%	g	% DMN	% DMN	%	g	% DMN	g	% DMN	g	% DMN
n-heptane			100.00	100.00%		93.36	153.35%	60.53%			6.64	153.35%	6.64	16.97%
2,6DMN	48.73	48.73%				13.02	21.38%	8.44%	34.79	100.00%	0.93	21.38%	35.71	91.30%
1,5DMN	8.54	8.54%				7.97	13.10%	5.17%	0.00	0.00%	0.57	13.10%	0.57	1.45%
1,6DMN	41.12	41.12%				38.39	63.06%	24.89%			2.73	63.06%	2.73	6.98%
Other	1.61	1.61%				1.50	2.47%	0.97%			0.11	2.47%	0.11	0.27%
Total DMN	100.00					60.88			34.79		4.33		39.12	
Overall Total			100.00			154.24					10.97		45.76	

n-heptane (g) 100

Crystallization yield 73.3%

Washing with n-heptane carried out at: 20 °C

	Washing		Sol. 2,6 DMN		5.8%		Solid		Wet/Solid		9.11%		Panel	
	g	%	g	% DMN	%	g	% DMN	g	% DMN	g	% DMN	g	% DMN	%
n-heptane	30.00	100.00%	34.19	521.34%	83.91%					2.45	521.34%	2.45	7.53%	7.01%
2,6DMN			3.38	51.56%	8.30%	32.09	100.0%	0.24	51.56%	32.33	99.31%	32.33	99.31%	92.34%
1,5DMN			0.53	8.07%	1.30%	0	0.0%	0.04	8.07%	0.04	0.12%	0.04	0.12%	0.11%
1,6DMN			2.55	38.85%	6.25%			0.18	38.85%	0.18	0.56%	0.18	0.56%	0.52%
Other			0.10	1.52%	0.24%			0.01	1.52%	0.01	0.02%	0.01	0.02%	0.02%
Total DMN			6.56			32.09		0.47		32.56		32.56		
Overall Total	30.00		40.74					2.92		35.01		35.01		

Overall n-heptane (g) 130.0

Crystallization yield 66.4%

Sol. 2,6 DMN means: solubility of solid 2,6-DMF in the solvent

TABLE B

Preparation of 2,6-dimethylnaphthalene having a high purity by crystallization with methanol
COMPARISON WITH EXAMPLE 1 OF EP 0792858

CRYSTALLIZATION CARRIED OUT AT : 20 °C

	Initial charge		% Methanol		Sol. 2,6 DMN		Mothers		Solid		Wet/Solid		Panel	
	g	% DMN	g	%	g	% DMN	% DMN	%	g	% DMN	g	% DMN	g	% DMN
MeOH			100.00	100.00%	92.11	166.76%	62.51%				7.89	166.76%	7.89	17.63%
2,6DMN	48.73	48.73%			8.01	14.50%	5.44%	40.03	100.00%		0.69	14.50%	40.72	90.98%
1,5DMN	8.54	8.54%			7.87	14.24%	5.34%	0.00	0.00%		0.67	14.24%	0.67	1.51%
1,6DMN	41.12	41.12%			37.88	68.57%	25.71%				3.24	68.57%	3.24	7.25%
Other	1.61	1.61%			1.48	2.68%	1.01%				0.13	2.68%	0.13	0.28%
Total DMN	100.00				55.23			40.03			4.73		44.77	
Overall total			100.00		147.34						12.62		52.66	

Methanol (g) 100 Crystallization yield 83.6%

Washing with methanol carried out at : 20 °C

	Washing		Sol. 2,6 DMN		Washing liquid		Solid		Wet/Solid		Panel	
	g	%	g	% DMN	g	% DMN	g	% DMN	g	% DMN	g	% DMN
MeOH	42.00	100.00%	46.66	918.45%	90.18%			3.23	918.45%	3.23	8.14%	7.53%
2,6DMN			1.30	25.52%	2.51%	39.33	100.0%	0.09	25.52%	39.42	99.34%	91.86%
1,5DMN			0.63	12.41%	1.22%	0	0.0%	0.04	12.41%	0.04	0.11%	0.10%
1,6DMN			3.03	59.73%	5.87%			0.21	59.73%	0.21	0.53%	0.49%
Other			0.12	2.34%	0.23%			0.01	2.34%	0.01	0.02%	0.02%
Total DMN			5.08			39.33		0.35		39.69		
Overall Total	42.00		51.74					3.58		42.92		

Overall Methanol (g) 142.0 Crystallization yield 80.9%

DELTA LOSS -43.2%

Sol. 2,6 DMN means: solubility of solid 2,6-DMF in the solvent